



AFRL-RZ-WP-TP-2008-2077

**TEMPERATURE MEASUREMENTS IN REACTING
FLOWS USING TIME-RESOLVED FEMTOSECOND
COHERENT ANTI-STOKES RAMAN SCATTERING (fs-
CARS) SPECTROSCOPY (POSTPRINT)**

Sukesh Roy, Paul J. Kinnius, Robert P. Lucht, and James R. Gord

**Engine Integration and Assessment Branch
Turbine Engine Division**

**AUGUST 2007
Interim Report**

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REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YY) August 2007		2. REPORT TYPE Journal Article Postprint		3. DATES COVERED (From - To) 08 August 2005 – 08 August 2007	
4. TITLE AND SUBTITLE TEMPERATURE MEASUREMENTS IN REACTING FLOWS USING TIME-RESOLVED FEMTOSECOND COHERENT ANTI-STOKES RAMAN SCATTERING (fs-CARS) SPECTROSCOPY (POSTPRINT)				5a. CONTRACT NUMBER IN HOUSE	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER 62203F	
6. AUTHOR(S) Sukesh Roy (Innovative Scientific Solutions, Inc.) Paul J. Kinnius and Robert P. Lucht, (Purdue University) James R. Gord (Engine Integration and Assessment Branch, Turbine Engine Division (AFRL/RZTC))				5d. PROJECT NUMBER 3048	
				5e. TASK NUMBER 04	
				5f. WORK UNIT NUMBER 304804AD	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> Propulsion Sciences Branch Aerospace Propulsion Division Air Force Research Laboratory, Propulsion Directorate Wright-Patterson Air Force Base, OH 45433-7251 Air Force Materiel Command, United States Air Force </div> <div style="width: 45%;"> Innovative Scientific Solutions, Inc. Purdue University </div> </div>				8. PERFORMING ORGANIZATION REPORT NUMBER AFRL-RZ-WP-TP-2008-2077	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Research Laboratory Propulsion Directorate Wright-Patterson Air Force Base, OH 45433-7251 Air Force Materiel Command United States Air Force				10. SPONSORING/MONITORING AGENCY ACRONYM(S) AFRL/RZTC	
				11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S) AFRL-RZ-WP-TP-2008-2077	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.					
13. SUPPLEMENTARY NOTES PAO case number AFRL/WS07-1603, 10 July 2007. ©2007 Elsevier B.V. The U.S. Government is joint author of the work and has the right to use, modify, reproduce, release, perform, display, or disclose the work. Published in Optics Communications, 281 (2008) 319-325. Document contains color.					
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15. SUBJECT TERMS CARS, fs-CARS, temperature, laser diagnostics					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT: SAR	18. NUMBER OF PAGES 14	19a. NAME OF RESPONSIBLE PERSON (Monitor) Amy Lynch 19b. TELEPHONE NUMBER (Include Area Code) (937) 255-6814
a. REPORT Unclassified	b. ABSTRACT Unclassified	c. THIS PAGE Unclassified			

Temperature measurements in reacting flows by time-resolved femtosecond coherent anti-Stokes Raman scattering (fs-CARS) spectroscopy

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Received 8 August 2007; received in revised form 11 September 2007; accepted 14 September 2007

Abstract

Time-resolved femtosecond coherent anti-Stokes Raman scattering (fs-CARS) spectroscopy of the nitrogen molecule is used for the measurement of temperature in atmospheric-pressure, near-adiabatic, hydrogen-air diffusion flames. The initial frequency-spread dephasing rate of the Raman coherence induced by the ultrafast (~ 85 fs) Stokes and pump beams is used as a measure of gas-phase temperature. This initial frequency-spread dephasing rate of the Raman coherence is completely independent of collisions and depends only on the frequency spread of the Raman transitions at different temperatures. A simple theoretical model based on the assumption of impulsive excitation of Raman coherence is used to extract temperatures from time-resolved fs-CARS experimental signals. The extracted temperatures from fs-CARS signals are in excellent agreement with the theoretical temperatures calculated from an adiabatic equilibrium calculation. The estimated absolute accuracy and the precision of the measurement technique are found to be ± 40 K and ± 50 K, respectively, over the temperature range 1500–2500 K.

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PACS: 78.47.p; 39.30.+w; 42.65.Dr; 82.53.Kp; 33.20.Fb

1. Introduction

Coherent anti-Stokes Raman scattering (CARS) spectroscopy of nitrogen and hydrogen using nanosecond (ns) lasers is widely employed for gas-phase temperature and species-concentration measurements [1,2]. The nonlinear interaction of the laser beams in ns-CARS generates a coherent nonresonant four-wave-mixing (FWM) signal along with the resonant CARS signal. Interpretation of the signal is complicated by the interference between the resonant and nonresonant signals. The nonresonant signal sometimes limits the accuracy and degrades the sensitivity of the technique [3]. Moreover, these measurements are

generally performed at low repetition rates (generally 10–20 Hz) because of the unavailability of high-repetition-rate, high-power ns lasers. This lack of temporal resolution hinders the study of unsteady phenomena in reacting flows.

The use of femtosecond (fs) laser systems for CARS spectroscopy has three significant potential advantages: (1) reduction or elimination of the nonresonant contribution to the CARS signal when the probe beam is delayed with respect to the pump beam, (2) reduction or elimination of the effects of collisions on the CARS signal, thereby reducing modeling uncertainty and increasing signal-to-noise ratio, and (3) the capability of generating signals at rates of 1 kHz or greater. The reduction or elimination of the nonresonant background and collisional effects will greatly simplify the modeling of CARS spectra and improve accuracy by eliminating the need for information

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concerning Raman linewidths. The advent of high-fidelity picoseconds (ps) and fs lasers has revolutionized the field of optical spectroscopy and enabled the development of tabletop systems for laser machining, generation of X-rays, extreme ultraviolet light, and attosecond optical pulses [4–8].

Schematic diagrams of the CARS process and the Raman-excitation processes with ns- and fs-lasers are shown in Fig. 1. In CARS, the wavelengths of the pump and Stokes beams are chosen such that the pump-Stokes frequency difference matches a Raman resonance frequency of the molecule. For the nitrogen molecule, the Raman resonance frequency for the $v'' = 0$ to $v' = 1$ band is approximately 2330 cm^{-1} . This excitation process creates a Raman coherence in the medium, resulting in a shifted signal when the medium is probed by another laser beam. In multiplex CARS using ns lasers, a narrowband pump beam and a broadband Stokes beam are employed for simultaneous excitation of numerous transitions in the ro-vibrational Raman band of the molecule. As shown in Fig. 1, in ns-CARS each pair of pump and Stokes frequencies is resonant with only one transition. For fs-CARS, however, multiple pump-Stokes pairs contribute to the excitation of the same transition, thereby creating a significant Raman coherence in the medium [9,10] despite the large frequency bandwidths of the pump and Stokes beams. Numerous Raman transitions are excited with the same phase when the pump and Stokes beams are nearly Fourier transform-limited. This in-phase impulsive excitation creates a very large coherence in the medium, which then decays as a result of the slight frequency differences between the neighboring transitions. This decay rate can be used to determine the temperature [11].

Time-resolved fs-CARS has been used for the first time by Leonhardt et al. to study the molecular beat phenomena in liquid phase benzene, cyclohexane, and pyridine [12]. Hayden and Chandler [13] first used fs-CARS for investigating the molecular vibrational dynamics of ground-state gas-phase benzene and 1,3,5-hexatriene. Schmitt et al. [14] used fs-CARS to study the ground- and excited-electronic-state dynamics of iodine vapor. These studies demon-

strated the potential for applying broad-bandwidth fs-lasers for gas-phase spectroscopic studies. Previously, the application of these lasers was thought to be of little value because of the excitation of multiple transitions by the broad laser pulses and the relatively inefficient coupling of these broad pulses to an individual transition as compared to the narrowband pulses more closely matched to the linewidth of these transitions. Hayden and Chandler [13] examined the dephasing of the coherence established by the pump and Stokes beams during the first few ps after the initial excitation and observed markedly different decay rates for gas-phase benzene and 1,3,5-hexatriene. They observed that the decay rate of the initial coherence was much faster for 1,3,5-hexatriene than for benzene due to dephasing of the initial orientation created by the laser pulses. Unlike benzene, in 1,3,5-hexatriene the laser pulses create a superposition of spatially oriented, vibrationally excited, rotational states that starts to change from the initial orientation due to the variation of rotational speeds of different molecules in the ensemble. Recently our group recognized that this initial dephasing rate during the first few ps can be used as a measure of gas-phase temperature, independent of any collisional influence [10,11].

The use of fs lasers for investigating the ultrafast dynamics of isolated molecule was pioneered by Scherer et al. [15]. The work of Dantus et al. [16] for observing the molecular vibration and rotational dynamics was also ground breaking in this regard. The work of Zewail's group [15,16] was based on pump-probe techniques where they prepared an excited state with a pump beam and detected the laser-induced fluorescence signal when excited by a delayed probed beam; this is similar to the fs-CARS technique, where the molecular coherence is prepared by the overlapping pump and Stokes beam and is then probed by a delayed probe beam. The review paper by Dantus [17] provides a comprehensive discussion of the coherent nonlinear spectroscopy based on ultrafast lasers.

Lang et al. [18] used fs-CARS of the H_2 molecule for determining molecular parameters and gas-phase temperature from the time-resolved oscillatory pattern of the Raman coherence following pump-Stokes excitation.

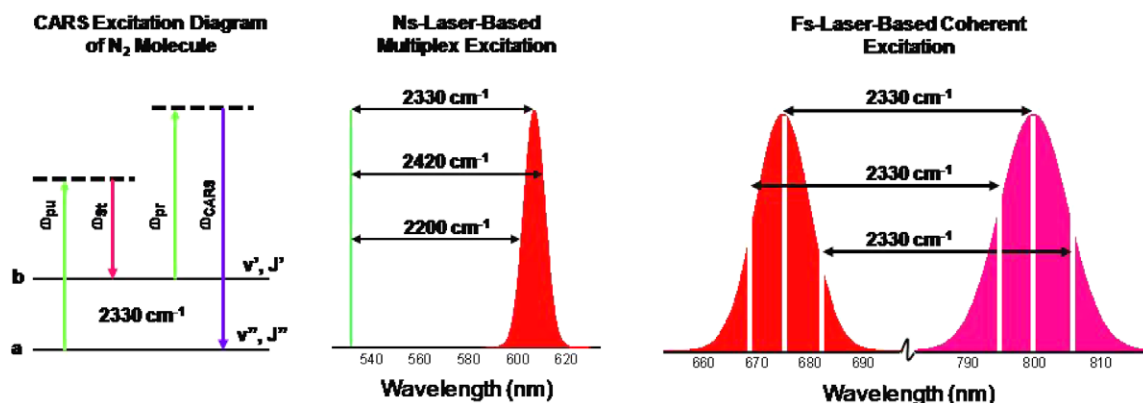


Fig. 1. Coherent excitation process in ns- and fs-laser-based CARS spectroscopy.

Those parameters were determined from the width and the relative heights of the recurrence peaks. Lang et al. [19] also used the time-resolved N_2 CARS signal for measuring temperature by focusing on recurrence peaks at ~ 320 ps or longer; on this time-scale, understanding of the collisional environment and the relaxation processes is essential for accurate temperature measurements. fs-CARS has also been used for measurements in a dense medium to investigate rotational energy-transfer processes [20], for determining the concentration of *ortho*- and *para*-deuterium [21], and for measuring single-shot temperature by probing the hydrogen molecule using a chirped probe pulse [22]. More recently, fs-CARS has been used for the detection of bacterial spores in the presence of other molecules [23], for the characterization of polymer thin films [24], and for background-free analysis of analytes trapped in aerogel pores [25].

The objective of the current study is to apply the time-resolved fs-CARS technique for temperature measurements in high-temperature flames, based on the frequency-spread dephasing rate after the initial impulsive excitation of the Raman coherence in the N_2 molecule by fs pump and Stokes beams. In this study, temperature from the time-resolved N_2 CARS signal is extracted by means of a simple theoretical model by concentrating on the signal decay during the first few ps after the pump-Stokes excitation. These decay results from the slight frequency mismatches between the neighboring Q-branch transitions and is completely insensitive to collisions [11]. The accuracy and precision of the measurement technique are also addressed.

2. Experimental system

A schematic diagram of the experimental system is shown in Fig. 2. The output of a 1-mJ, 1-kHz, 85-fs, Ti:Sapphire regenerative amplifier (Model: Libra, Coherent, Inc.) at 800 nm is used to pump an optical parametric amplifier (OPA). Approximately 15% of the energy of the regenerative amplifier is used as the Stokes beam, and the remainder is used to pump the OPA. The laser beam from the frequency-doubled OPA is centered at ~ 675 nm, with an approximate energy of 25 μ J/pulse. This beam is then split equally to yield the pump and probe beams for the CARS signal generation. The full-width-at-half-maxima (FWHM) of the frequency spectra of the pump (probe) and Stokes lasers were approximately 160 cm^{-1} and 220 cm^{-1} , respectively. The timing of the probe beam with respect to the pump and Stokes beams was varied for acquisition of the probe-delay scans using a motorized translation stage. A short-pass filter was used to block the scattered light from the pump and probe beams, and the CARS signal centered at 584 nm was detected with a low-noise, high-bandwidth photodetector (10-MHz Adjustable Photoreceiver, Model: 2051, New Focus). The signal was acquired using a lock-in amplifier, and each data point was averaged over 300 laser shots. Measurements were performed in an atmospheric-pressure, near-adiabatic hydrogen-air flame stabilized over a Hencken burner. Temperature was varied by changing the equivalence ratio (ϕ) of the flame (equivalence ratio is defined as the ratio of actual fuel-to-air over fuel-to-air for the stoichiometric condition [26]). The temperature of the flame stabilized

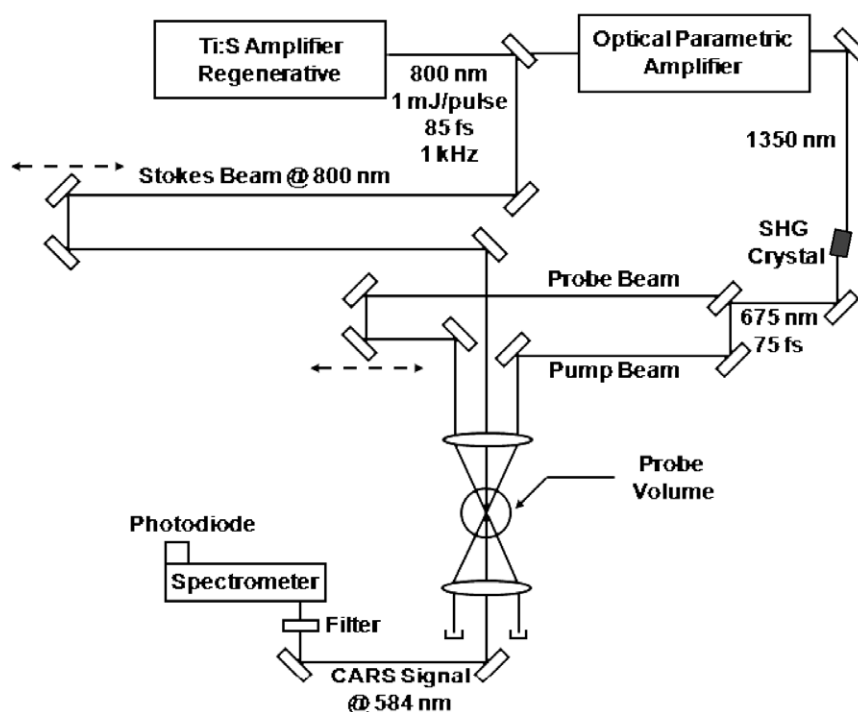


Fig. 2. Schematic diagram of fs-CARS system.

over the Hencken burner increases with equivalence ratio for $\phi \leq 1.0$, after which it decreases due to incomplete combustion of the fuel [27].

3. Results and discussion

Time-resolved fs-CARS signals of nitrogen as a function of probe pulse delay with respect to the pump beam at various equivalence ratios are shown in Fig. 3. The decay of the Raman coherence during the first few ps after the initial impulsive excitation by the nearly transform-limited pump and Stokes laser pulses is the focus of our experiments. The effects of collisions on this time scale are not important for pressures less than 10 bar. It is evident from the figure that the rate at which the signal decays increases with flame equivalence ratio and temperature. The signal decays faster with increasing temperature because of the contributions of more transitions over a wider frequency range to the initial Raman coherence. This results from redistribution of the population to higher energy levels with increasing temperature. The spectrally broad, nearly transform-limited pump and Stokes pulses simultaneously excite all the transitions accessible within the bandwidth, frequency pairs of the lasers. The resulting coherence decay thereafter as a result of the slight frequency mismatches between neighboring transitions. The signal reaches a maximum value at a probe delay of zero because of the contribution from both resonant and nonresonant signals. In Fig. 3 all signals were normalized to a peak intensity of 100 at time zero. The oscillatory behavior of the signal is qualitatively similar for temperatures in the range 1600–2400 K. The characteristic frequency of the oscillations observed in Fig. 3 is approximately 900 GHz, which corresponds to the beat frequency between the $v' = 1 \rightarrow v'' = 0$ and $v' = 2 \rightarrow v'' = 1$ vibrational bands and also between the $v' = 2 \rightarrow v'' = 1$ and $v' = 3 \rightarrow v'' = 2$ vibrational bands.

Theoretical spectra of N_2 CARS for three different temperatures are shown in Fig. 4. These spectra were calculated using the Sandia CARSFT code [28]. For

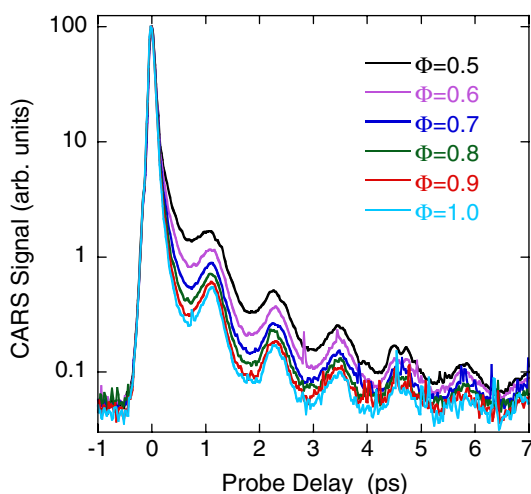


Fig. 3. Time-resolved fs-CARS signal for various equivalence ratios (ϕ).

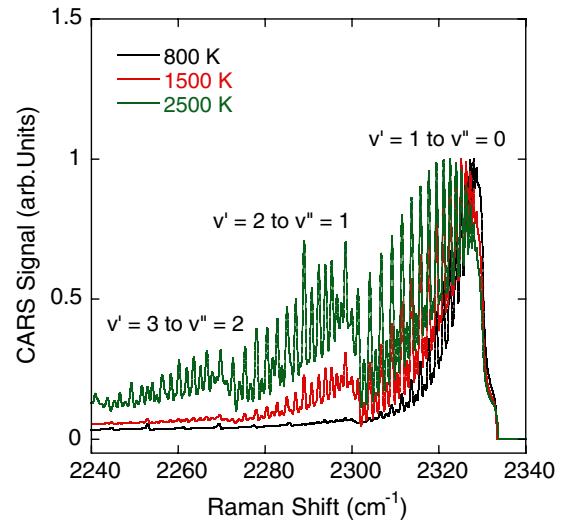


Fig. 4. Theoretical CARS spectra of nitrogen calculated using Sandia CARSFT code [26].

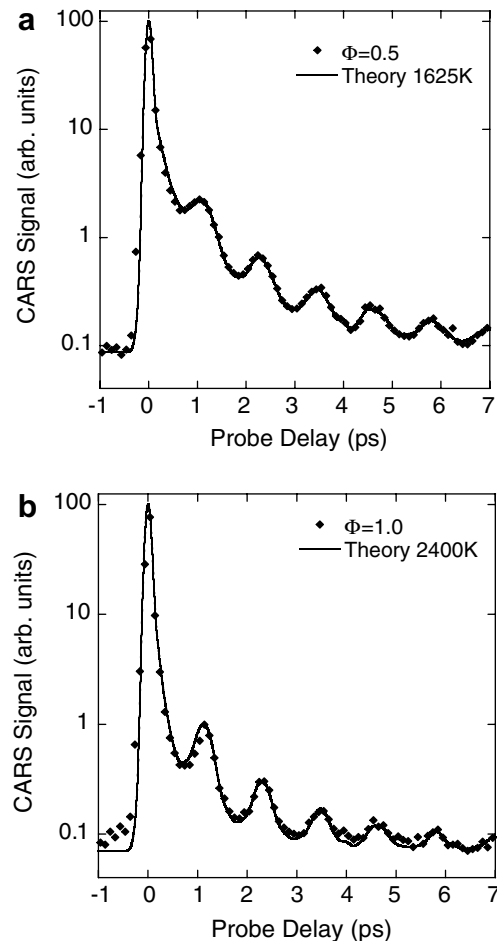


Fig. 5. Comparison of theoretical and experimental time-resolved fs-CARS signals for (a) $\phi = 0.5$ and (b) $\phi = 1.0$. Theoretical signals were calculated using Eq. (1). The solid lines represent the best-fit theoretical signal to the experimental data points and is determined by least-square fitting to the experimental data points.

temperatures less than 800 K, only the first vibrational band appears, which results in a time-resolved signal without oscillations during the first 10 ps after the initial excitation, as shown in our previous work [11]. The first hot-band ($v' = 2 \rightarrow v'' = 1$) begins to appear for temperatures greater than 1000 K, and the second hot-band ($v' = 3 \rightarrow v'' = 2$) appears at approximately 2000 K. The frequency difference between the band heads is approximately 30 cm^{-1} , as is evident in Fig. 4. This 30-cm^{-1} frequency difference corresponds to a 900 GHz in beat frequency.

Fig. 5 shows the fit between the theoretical and experimental probe-delay scans for $\phi = 0.5$ and $\phi = 1.0$. The best-fit theoretical probe-delay scans are used to extract temperatures from experimental scans. The theoretical modeling is discussed in detail by Lucht et al. [11]. The theoretical signal as a function of probe delay is calculated using the following equation.

$$S(\tau) = \int_{-\infty}^{+\infty} I_{\text{pr}}(t - \tau) [P_{\text{res}}(t) + P_{\text{nres}}(t)]^2 dt + \text{Background noise} \quad (1)$$

where the resonant signal (P_{res}) is calculated as

$$P_{\text{res}}(t) = \beta \left[\int_{-\infty}^t E_p(t') E_s(t') dt' \right] \times \sum_i \left\{ \Delta N_i \left(\frac{d\sigma}{d\Omega} \right)_i \cos(\omega_i t) \exp(-\Gamma_i t) \right\} \quad (2)$$

In Eq. (2), $E_p(t)$ and $E_s(t')$ are the time-dependent electric fields of the pump and Stokes laser fields, respectively, ΔN_i is the difference in population between the excited and ground levels of a transition, $\left(\frac{d\sigma}{d\Omega} \right)_i$ is the Raman cross section (for a particular transition i), ω_i is the frequency of the Raman transition, Γ_i is the coherence dephasing rate due to collisions, which will only be significant for longer time scales or higher pressures, and β is a scaling factor used to match the experimental signal with the theoretical spectrum. In Eq. (1), I_{pr} is the intensity of the probe beam, and P_{nres} is the nonresonant polarization calculated as

$$P_{\text{nres}}(t) = \alpha E_p(t) E_s(t) \quad (3)$$

where α is a nonresonant scaling factor. For each experimental scan, the ratio α/β is varied to match the theoretical

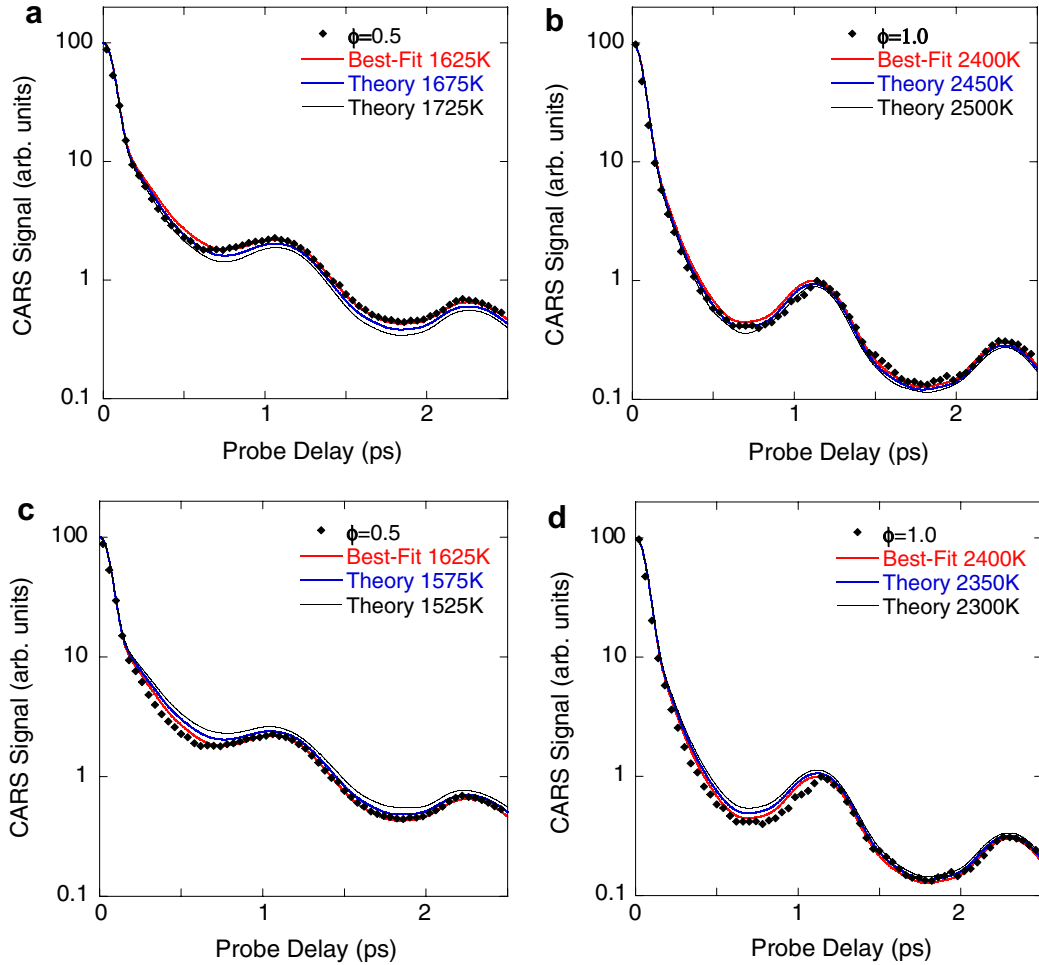


Fig. 6. Comparison of theoretical and experimental scans when the best-fit (based on least-square fit) temperature values were changed by $\pm 50 \text{ K}$ and $\pm 100 \text{ K}$ for $\phi = 0.5$ and $\phi = 1.0$. Scans are expanded versions of those shown in Fig. 5. Fig. 6a and b show probe-delay scans $\phi = 0.5$ and $\phi = 1.0$, respectively, when temperatures are varied by $+50 \text{ K}$ and $+100 \text{ K}$. Fig. 6(c) and (d) show probe-delays scans for $\phi = 0.5$ and $\phi = 1.0$, respectively, when temperatures are varied by -50 K and -100 K .

and experimental signal-decay rates. For the two equivalence ratios shown in Fig. 5, adiabatic flame temperatures are calculated to be 1642 K and 2382 K for $\phi = 0.5$ and $\phi = 1.0$, respectively, [26]. These measurements show very good agreement between the adiabatic flame temperatures and the temperatures extracted from the experimental scans by least-square fitting with the calculated theoretical time-resolved signal using Eq. (1). In Fig. 5, solid lines represent the theoretical best-fit signal and symbols represent the experimental data points.

To address the temperature sensitivity of the measurements, experimental scans shown in Fig. 5 were compared with theoretical scans by varying the best-fit temperature by ± 50 K and ± 100 K as shown in Fig. 6. Fig. 6a and b show the expanded versions of the time-resolved scans for $\phi = 0.5$ and $\phi = 1.0$ when the theoretical temperatures were varied by $+50$ K and $+100$ K from the best-fit values; Fig. 6c and d show the same scans when the theoretical temperatures were changed by -50 K and -100 K from the best-fit values. The clear mismatch between the experimental and theoretical signals for temperatures adjusted by 50 K and 100 K from the best-fit value shows the sensitivity of the current measurement technique is ± 50 K. Changing the temperature by ± 25 K did not yield any perceptible variation in the theoretical signal profiles.

To address the accuracy and precision of the proposed measurement technique, the temperature extracted from the experimental signals is compared with adiabatic flame temperatures, as shown in Fig. 7. The extracted temperatures are within 40 K of the adiabatic flame temperatures for equivalence ratios ranging from 0.5 to 1.0. The systematic error in the measurements cannot be evaluated quantitatively from these results, but appears to be much smaller than random errors of approximately ± 50 K. It might be expected that systematic errors in fs-CARS will be reduced compared to ns-CARS because collisions do not affect the

fs-CARS signal in the first few ps after impulsive pump-Stokes excitation. Consequently, no knowledge of Raman linewidths is required to model the fs-CARS signal behavior. For ns-CARS, on the other hand, accurate Raman linewidth data is required for temperature determination [29]. The three sets of data points shown in Fig. 7 were acquired on three different days spanning two weeks in time. The standard deviation of the extracted temperatures was approximately 50 K for an equivalence ratio of 1.0, but the standard deviation was approximately 11 K for $\phi = 0.5$ and $\phi = 0.8$. These measurements suggest the accuracy and precision of the measurement technique are within ± 40 K and ± 50 K, respectively.

4. Conclusions

Temperatures ranging from 1500 K to 2400 K is measured using the time-resolved fs-CARS technique in atmospheric-pressure, near-adiabatic, hydrogen-air diffusion flames. In this study, wavelengths of the pump and the Stokes beams are chosen to probe the vibrational transitions of the N_2 molecule. The initial decay of the Raman coherence results from the slight frequency mismatches among the transitions, excited by the broad pump and Stokes beams. The rate of decay of the initial Raman coherence was used as a measure of temperature. The fs-CARS signals within the first few ps after pump-Stokes excitation are free from collisional influence for pressures less than 10 bar and, therefore, do not require any knowledge of the collisional-relaxation processes for either temperature or concentration measurements from experimental signals. The temperatures extracted from the time-resolved signals agreed very well with the adiabatic flame temperatures for a range of equivalence ratios. The estimated accuracy and precision of the measurement technique are within ± 40 K and ± 50 K, respectively. The estimated systematic errors in the measurements are less than ± 50 K.

Acknowledgements

Funding for this research was provided by the Air Force Office of Scientific Research (Dr. Julian Tishkoff, Program Manager), by the Air Force Research Laboratory, Propulsion Directorate, Wright-Patterson Air Force Base under Contract No. F33615-03-D-2329; by the National Science Foundation, Combustion and Plasma Program under Award Number 0413623-CTS. The authors gratefully acknowledge the technical assistance of Mr. Kyle D. Frische.

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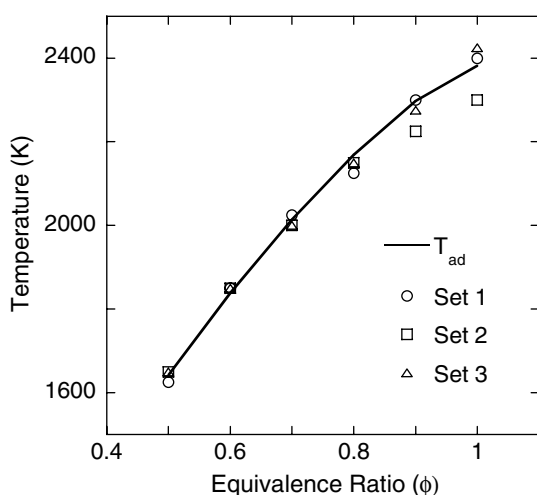


Fig. 7. Comparison of experimentally measured temperatures with adiabatic flame temperatures for atmospheric-pressure, near-adiabatic, hydrogen-air diffusion flame. Sets represent data acquired on three days over a two-week time period.

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